Nafion/Polyimide based programmable moisture-driven actuators for functional structures and robots

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Abstract:

Sustainable and environmentally friendly actuators powered by humidity, light and magnetic field are of great significance to facilitate application of microrobots. Among them, moisture-driven actuators have attracted a growing interest as a result of the widespread presence and ease of use of humidity. Nevertheless, the rapid acquisition and large-scale application of the moisture-driven actuators is still difficult since synthesis process of humidity-sensitive materials is time-consuming and complex. In this paper, we proposed a facile and rapid method to prepare double-layer moisture-driven actuators by integrating commercial humidity-sensitive Nafion ™ membrane and polyimide (PI) tape with good stability. Compared with other double-layer moisture-driven actuators, the so-obtained actuator has excellent performance in both the bending curvature (from -0.98 cm^{-1} to 5.34 cm^{-1}) and the response speed (0.29 cm^{-1}/s). Through programmable structure design, a series of functional structures with complex deformation modes, i.e. torsion and bending, were realized. By imitating organisms like birds, vines, inchworms and ants, a series of soft robots have been developed based on the programmable structures to achieve behaviors including grasping, crawling and weight-bearing. The proposed programmable moisture-driven actuators have shed light on the fast development of environmental-friendly functional microrobots making use of ambient humidity.

Key words: moisture-driven actuators, humidity-sensitive, Nafion, polyimide, programmable structure, functional microrobots

1.Introduction

Microrobots in centimeter scale with autonomous and continuous motion have considerable potential in the fields of healthcare, environment, and military applications [1]. Nevertheless, how to provide the energy supply for microrobots is still a serious challenge. Compared with on-board battery devices, using natural energy from the environment to drive microrobots is a more effective and environmentally friendly approach, for example using humidity, heat, light, magnetic field and so on [2-4]. In this aspect, a great deal of inspiration can be drawn from the flora and fauna of nature. For example, the scales of pine cones can open and close according to the change of humidity. Sunflowers can change the facing direction corresponding to the irradiation of the sun. Migratory birds can migrate seasonally according to the guidance of the geomagnetic field. Inspired by the stimulus-response mechanism of these organisms, many soft actuators that can deform under the stimulation of natural energy have been developed, including moisture-driven [5, 6], light-driven [7, 8], thermal-responsive [9, 10] and magnetic-responsive actuators [11, 12], which have been widely used in soft robots [13, 14], intelligent clothing [15, 16], and photoelectric control [17, 18]. Among them, moisture-driven actuators have attracted a growing interest due to their unique capability of the biomimetic movements and interfacing with biological environments since proposed in 1997 [19].

Moisture-driven actuators usually achieve deformation by utilizing the hygroscopic swelling properties of humidity-sensitive materials, such as graphene oxide (GO) [20-22], MXene [23, 24], cellulose [25, 26], chitosan [27, 28] and some synthetic polymer materials [29, 30]. However, the synthesis process of moisture-driven actuators using these materials is time-consuming and complex, which greatly limit the rapid acquisition and large-scale application of the moisture-driven actuators. Therefore, a facile and rapid preparing method with low cost is an urgent need. It seems to be an effective way to use some commercial humidity-sensitive materials to prepare moisture-driven actuators. For example, Mu et al. [31] prepared a kind of vapor actuator through selective forming method of spraying a layer of perfluorosulfonic acid (PFSA, also known as Nafion) solution on a PET inert substrate, which can realize 2D and 3D deformation. Compared with other synthetic humidity-sensitive materials, Nafion is not only easy to obtain, but also has good hygroscopic deformation ability. Hence in this work, a facile and rapid method was proposed by using commercial Nafion$_{TM}$ membrane as the humidity-sensitive layer and combined with polyimide (PI) tape to prepare a moisture-driven actuator.

In terms of the structure, moisture-driven actuators can be divided into single-layer actuators and double-layer actuators according to whether there is a restricted layer. It is understood that the deformation of single-layer moisture-driven actuator depends on the humidity gradient and the water molecules on the hygroscopic side will begin to migrate to the other side over time to make the water distribution uniform, leading to the actuator return to original shape. Compared with single-layer actuators, double-layer actuators can produce more stable and larger deformation. However, the bending deformation of the double-layer actuator only can no longer meet the complex action requirements of the moisture-driven actuators, and it is necessary to further enrich the deformation behavior through programmable structure design. At present, there are two kinds of methods to conduct programmable structure design for moisture-driven actuators, one is to control the structure uniformity of the humidity-sensitive layer to enable uneven deformation after moisture absorption [32,33]. The other is to control the two-dimensional shape of the hygroscopic surface to make a specific deformation of the whole actuator [34, 35]. In this work, through the programmable design on the surface of the humidity-sensitive layer, a series of functional structures have been obtained to realize various deformation, which are applied to different types of soft robots.

In this paper, we demonstrate a moisture-driven actuator that can be fabricated quickly and cost effectively by making use of a layer of commercial humidity-sensitive film - Nafion and a layer of humidity-insensitive polyimide (PI) adhesive tape. Firstly, the moisture response characteristics of single-layer Nafion actuator and its relationship with cation type and thickness were studied. Subsequently, a double-layer actuator was assembled, the performance of which was evaluated by changing the thickness, temperature and humidity during assembling. Furthermore, a series of complex deformations such as bending and torsion were realized for the double layer actuator through programmable structure design. Finally, the programmable structures were applied to different soft robots, including a cross-shaped gripper inspired by eagle claws, a spiral gripper inspired by vines, a crawling robot inspired by inchworms and a weight-bearing robot inspired by ants.

2.Results and Discussion

Nafion is one type of polymers with good hygroscopic ability, whose molecular formula is shown in Figure 1 a. The fluorocarbon long chains constitute the backbone of Nafion, and the short side chains are terminated by sulfonic acid groups, which are hydrophilic groups that can bind water molecules. Besides, the interior of the Nafion membrane is porous and connected with irregular nanochannels, as shown in Figure 1.b, which can accommodate a large number of water molecules. The water in the nanochannels can dissociate the cations at the end of the sulfonic acid group into hydrated cations that can freely migrate therein. After absorbing water, the polymer network of Nafion swells by a volume change, as shown in Figure 1.c, resulting in macroscopic deformation. The specific microscopic physical mechanism is depicted in Figure S1. When one side of the Nafion membrane absorbs moisture, the hygroscopic side swells first. Furthermore, the entrance of a large number of water molecules reduces the cation concentration of the hygroscopic side, so the cations of the back side migrate by carrying water molecules to the hygroscopic side under the concentration gradient, which causes the hygroscopic side to expand further and the back side to contract, thereby exhibiting a bending deformation toward the back side.

Due to the existence of substantial cations in the nanochannels inside the Nafion membrane, the humidity-sensitive capacity of cations will inevitably affect deformation characteristics of the actuator. In order to investigate the effects of cation types on the deformation of Nafion, the moisture response process of Nafion (thickness of 0.051 mm) with Li^+, Na^+ and K^+ were tested respectively. By blowing air with saturated humidity to one side of the actuator using a humidifier through a tube with a diameter of 1 cm, the deformation of actuators with different cations were recorded. Among them, the sample with Li^+ exhibits the best moisture-driven performance, which reaches a bending curvature of 2.15 cm^{-1} within 1 second as shown in Figure 1.d, where the curvature is defined as (Bending radian)/(Length of the actuator). While it takes 3 and 4 seconds to achieve the same deformation respectively for the samples with Na^+ and K^+ (Figure S2). The bending curvature of the three samples with time are described in Figure 1.e, in which the sample with Li^+ not only has the highest response speed, but also has the highest recovery speed, which can be attributed to the fact that Li^+ has the strongest ability to bind water molecules [36]. In addition, the effects of thicknesses on the deformation of single-layer Nafion actuator were investigated by taking Nafion membranes with different thicknesses as the samples (N-112 with a thickness of 0.051 mm, NE-1135 with a thickness of 0.089 mm and N-117 with a thickness of 0.183 mm, all exchanging Li^+), and the results are shown in Figure S3 and Figure 1 f. It can be seen that the sample with smaller thickness has larger bending curvature and higher response speed, which is related to the stiffness change caused by the thickness. Figure 1 g compares the performance of pure Nafion with other single-layer moisture-driven actuators (the best actuating performance of other materials from the references are selected, and the numbers of references are marked in parentheses). It is clear that Nafion has excellent performance both in terms of bending curvature and response speed.

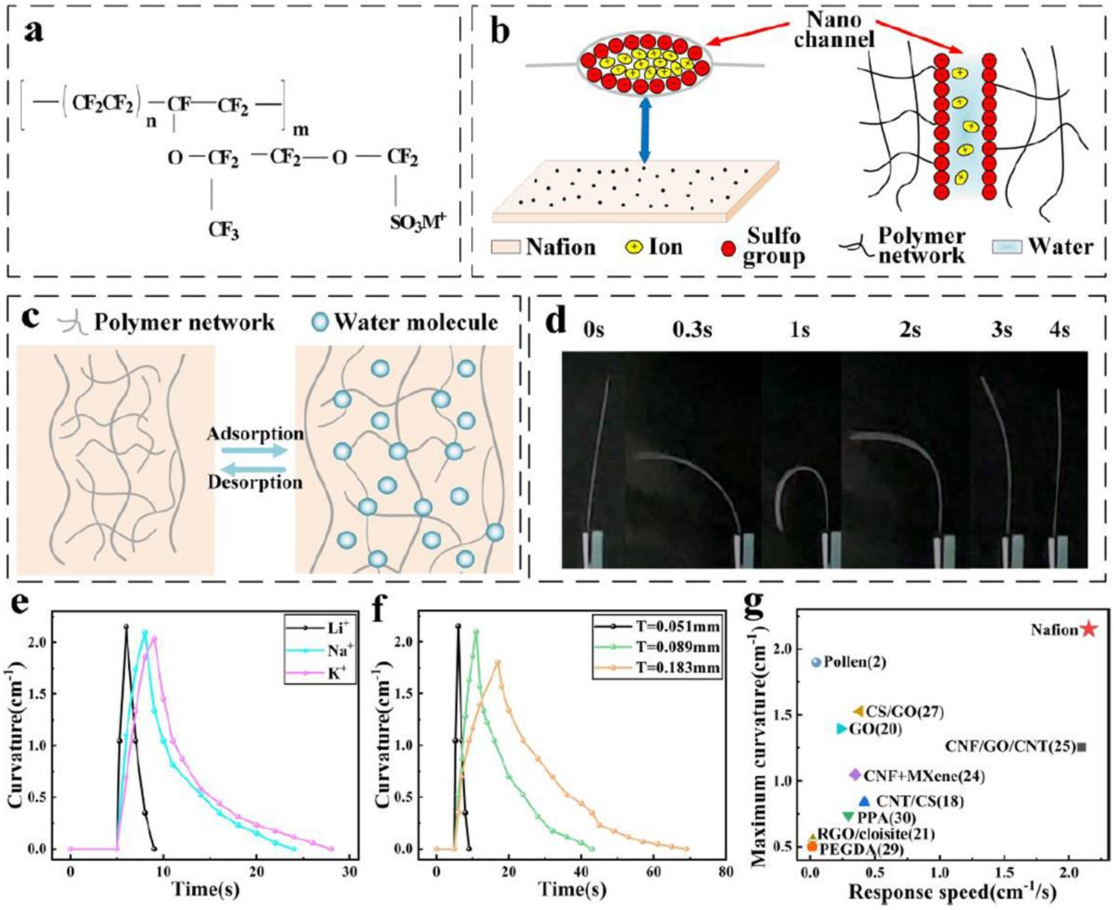

Figure 1. a. The molecular formula of Nafion. b. Schematic diagram of the microstructure of Nafion. c. The swelling of polymer network under the action of water molecules. d. Moisture response process of Nafion with Li^+. e. The curves of bending curvature with time of Nafion with different cations. f. The curves of bending curvature with time of Nafion with different thickness. g. Performance comparison the of pure Nafion with that of other single-layer moisture-driven actuators.

In view of the superior moisture absorption ability of Nafion, we chose it as the moisture sensitive layer to fabricate the bilayer actuator by connecting with a layer of humidity insensitive material. In this work, PI tape with good humidity insensitivity and thermal stability (RHL 12530#, thickness of 0.03 mm ± 0.01 mm, where the thickness of PI substrate is 0.0125 ± 0.005 mm and the thickness of adhesive layer is 0.0175 ± 0.005 mm) is selected as the binding layer of the double-layer actuator. Figure 2 a shows a very facile and rapid process to prepare a double-layer actuator. One side of the Nafion membrane was treat with sandblasting and then bond to the PI tape. Sandblasting process can effectively improve the surface roughness of Nafion, thus enhancing the adhesion between Nafion layer and PI tape layer. Figure 2 b shows that sand blasting can increase the adhesion between layers from 315.7 kPa to 350.2 kPa. The appearance of the prepared double-layer actuator is shown in Figure 2 c. As can be seen from the cross section observed by the electron microscope (Figure 2 d), the connection between the layers is very tight and there are no gaps (the thickness of the PI tape is 0.026 mm and the thickness of Nafion is 0.183 mm), which ensures good working performance of the Nafion/PI actuator. The working mechanism of the Nafion/PI moisture-driven actuator is shown in Figure 2 e. When the humidity is higher than the assembly humidity (relative humidity where the actuator is assembled), the Nafion layer swells and then bends to the PI layer side, while when the humidity is lower than the assembly humidity, the Nafion layer shrinks and then bends to the spontaneous side, resulting in bidirectional deformation. Figure 2 f shows the deforming behavior of the prepared double-layer actuator on different surfaces, which remains flat on dry gloves while curls on wet hands.

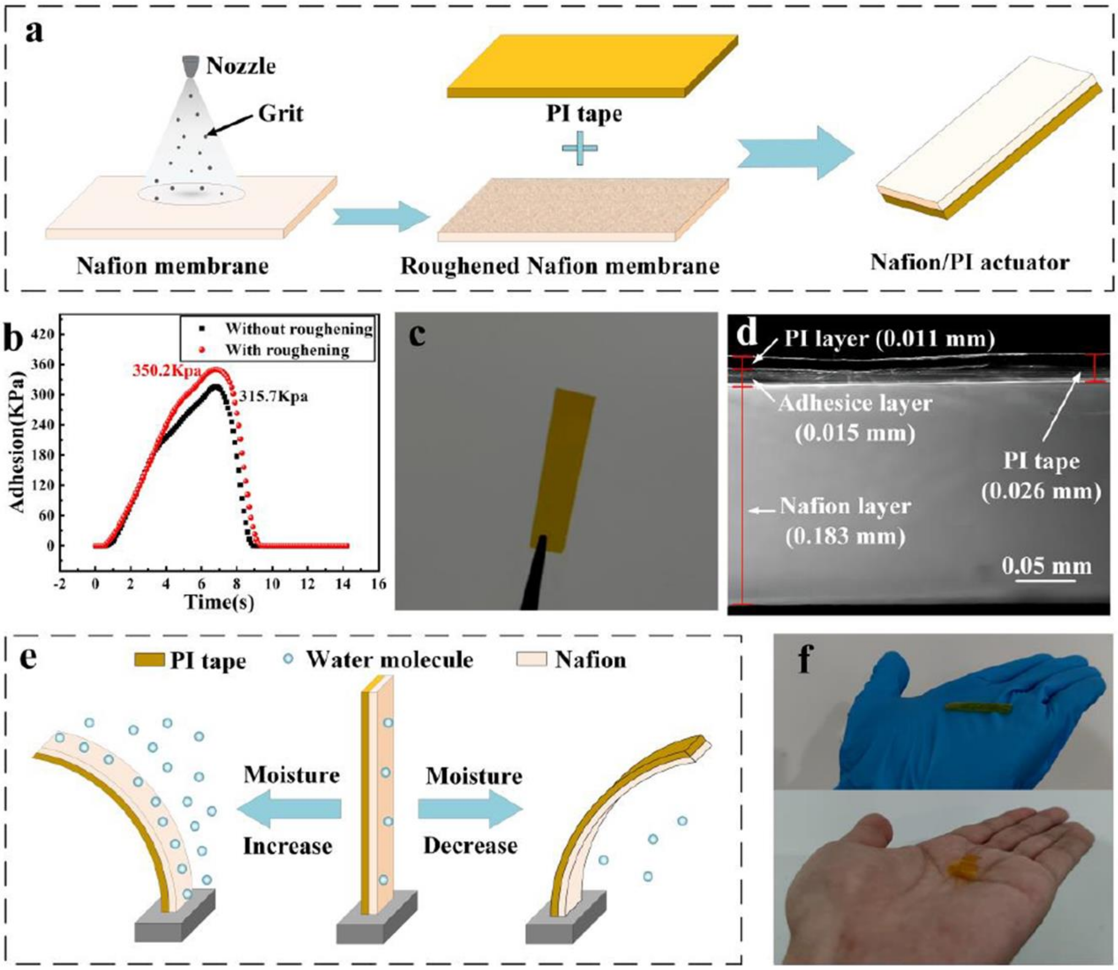

Figure 2. a. Preparing process of Nafion/PI actuator. b. The effect of roughening on the adhesion between layers. c. The appearance of the prepared double-layer actuator. d. The cross section of the actuator observed by the microscope. e. The working mechanism of the Nafion/PI moisturedriven actuator. f. The deforming behavior of the prepared on different surfaces.

In order to illustrate the performance of the prepared double-layer actuator, the bending curvature of the actuator was tested in different environments with various humidity. Figure 3 a show the bending deformation of the double-layer actuator with a Nafion layer of 0.051 mm (the size of the cantilever structure is 5 mm × 25 mm) in environments with different stable humidity. When the humidity is higher than 50%, the actuator bends to the Nafion side, while when the humidity is less than 30%, the actuator bends to the PI side, indicating that the actuator can achieve bidirectional deformation. The effects of thickness on the performance of the double-layer actuator were also investigated. Figure S4 shows the deformation of the double-layer actuator with 0.089 and 0.183 mm Nafion layers. It can be seen that the deformation of the actuator decreases as thickness of Nafion layer increases. Figure 3 b compares the bending curvature of double-layer actuators with of Nafion layers of different thickness. As the thickness increases, the bending curvature shows a decreasing trend. In addition, the bending curvature range of the bidirectional deformation is also related to assembly humidity. Figure S5 shows the bending deformation of the double-layer actuator assembled in 70% RH. When the humidity is higher than 70%, the actuator bends to the Nafion side, while when the humidity is less than 70%, the actuator bends to the PI side. Figure 3 c compares the range of bending curvature for the actuators assembled at 40% RH and 70% RH, respectively. It can be seen that the range of deformation appears to shift only along the ordinate without a change in margin. The switching point from the negative angle to the positive angle is determined by the assembly humidity, which means that we can program the deformation range of the double-layer actuator by changing the assembly humidity, which we call state programming. To demonstrate the stability of the prepared double-layer actuator, the sample with a 0.051 mm Nafion layer were tested between 22% RH and 74% RH for 100 cycles, and the results are shown in Figure 3 d. It can be concluded that the deformation range of the prepared humidity actuator does not change significantly with the increase of operation duration, indicating a good stability. Since temperature also affects the ambient humidity, the deformation of the actuator was tested when exposed on water with different temperatures, as shown in Figure 3 e. As the temperature increases from 20 °C to 80 °C, the bending curvature of the actuator also increases from 3.14 cm^{-1} to 4.71 cm^{-1}. This is because the increase in temperature with lead higher saturation water vapor pressure, and more water molecules can be accommodated in the air, so that the actuator can absorb more water molecules to achieve greater deformation. Figure 3 f depicts the variation of the bending curvature of the actuator with temperature, there is an approximately linear relationship between them.

Moreover, the response speed of actuators with Nafion layers with different thickness were also tested (the method is the same as the single-layer actuator). It can be identified from Figure 3 g that the actuator with thinner Nafion layer has a faster response speed and recovery speed. This is because the increase in the thickness of the moisture sensitive layer results in the need to absorb more water molecules to achieve the same deformation. By comparing with the performance of other double-layer actuators in Figure 3 h (the best actuating performance of other double-layer actuators from the references are selected, and the numbers of references are marked in parentheses), it can be seen that the prepared Nafion/PI moisture-driven actuator has excellent performance in both the maximum bending range and the response speed. On the one hand, the hydrophilic groups in most humidity-sensitive materials are hydroxyl groups, such as graphene oxide, cellulose and chitosan [20, 25, 27], which are connected with water molecules through hydrogen bonds. While in Nafion, it is the sulfonic acid group that has the hygroscopic property, which is mainly combined with water molecules through ionic bonds. Since the hydrophilicity of ionic bonds is stronger than that of hydrogen bonds, the water absorption capacity of sulfonic acid groups is stronger than that of hydroxyl groups, which endows the double-layer actuator with high response speed. On the other hand, the cations in Nafion like Li^+ are also hygroscopic and can combine with multiple water molecules to form hydrated cations [36], which greatly increases the number of absorbed water molecules. In addition, there are a large number of nano channels for water molecules transmission inside Nafion [31], which can accommodate a large number of water molecules. Therefore, the double-layer actuator with Nafion has a larger hygroscopic deformation compared with actuators based on other humidity sensitive materials.

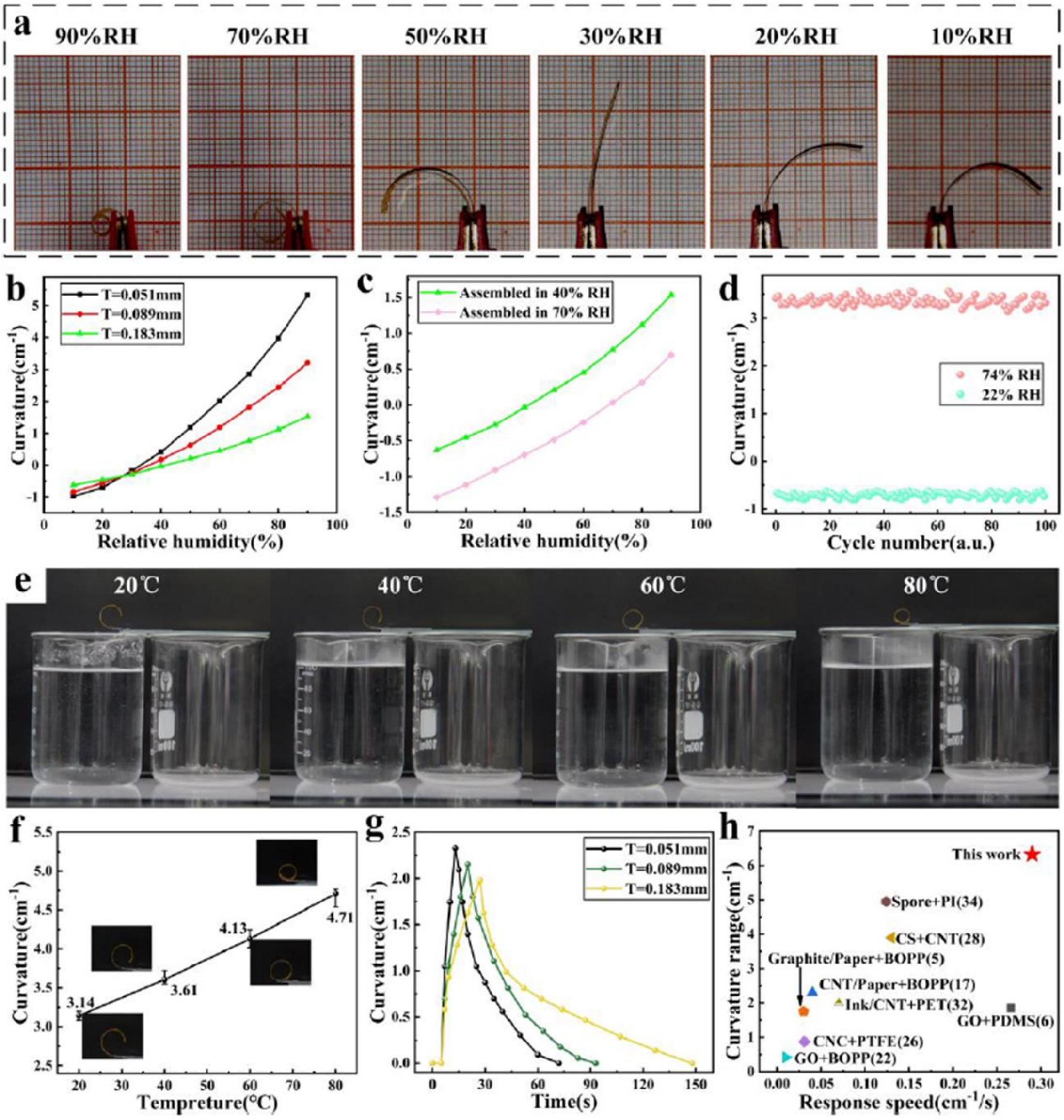

Figure 3. a. The bending deformation of the double-layer actuator with a Nafion layer of 0.051 mm in environments with different stable humidity. b. The bending curvature of double-layer actuators with of Nafion layers of different thickness. c. The bending curvature of the actuators assembled at 40% and 70% humidity. d. The stability of the prepared double-layer actuator. e. The deformation of the actuator exposed to water at different temperatures. f. The variation of the bending curvature of the actuator with temperature. g. The curves of bending curvature with time of actuators with Nafion layers of different thickness. h. Performance comparison of the prepared actuator with that of other double-layer moisture-driven actuators.

In order to achieve more complex deformation of the prepared double-layer actuator, the programmable structure is designed by mask pasting. As shown in Figure 4 a, the PI strip tape is attached to surface on the moisture-absorbing side of the double-layer actuator at the same interval. At this time, only the exposed Nafion surface can absorb moisture and deform, while the area covered by PI tape cannot actively deform. Therefore, samples cut in different directions can achieve different actions due to uneven deformation. The samples cut according to the four different ways of 1-4 can achieve four kinds of deformation, including left rotation torsion, right rotation torsion, bending along the length direction and bending along the thickness direction, as shown in Figures 4 b-e, respectively. In addition, other structural deformations can be achieved by designing the hygroscopic surface. For example, "S" type deformation can be achieved by sticking two strips of PI tape asymmetrically on both sides of the Nafion (Figures 4 f). While sticking two discontinuous PI tapes on the same side of Nafion can realize the "U" type deformation of rotating around a fixed point (Figures 4 g), which is similar to the revolute pair in the mechanical hinge structure.

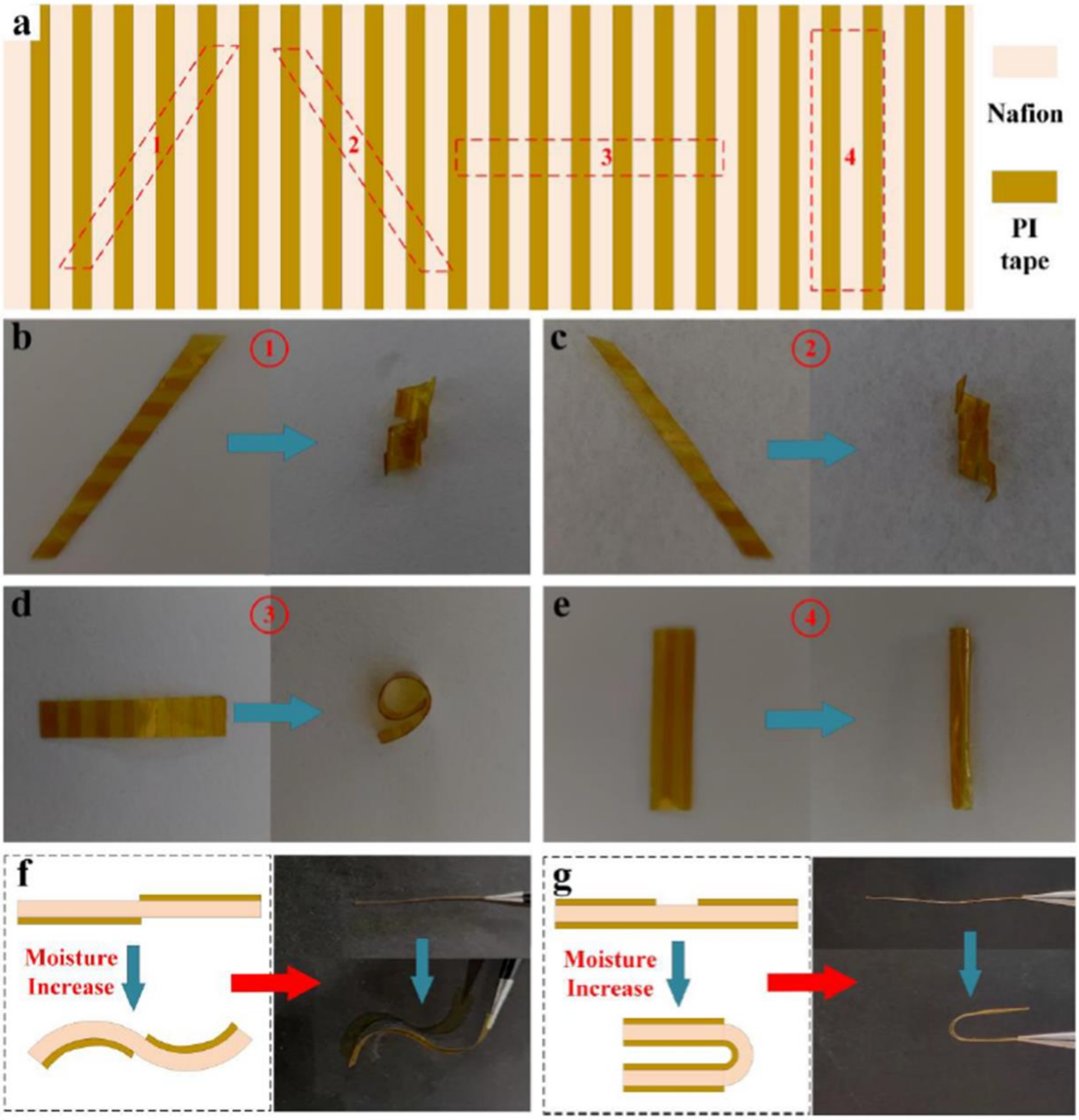

Figure 4. a. The PI strip tape is attached to surface on the moisture-absorbing side of the double-layer actuator at the same interval. b. Deformation of left rotation torsion. c. Deformation of right rotation torsion. d. Deformation of bending along the length direction. e. Deformation of bending along the thickness direction. f. "S" type deformation of the actuator. g. "U" type deformation of the actuator.

Based different programmable structures, we further developed a series of bionic soft robots with different functions. Inspired by the claws of birds, we designed a cross-shaped flexible gripper that can grasp blocky objects, as shown in Figure 5 a. A typical grasping process is shown in Video S1 and Figure 5 b, where the mass of the gripper is 26 mg and that of the grasped foam is 28 mg. First, the gripper is close to the foam and bends under high humidity to grip the foam. Then the foam is transferred from container 1 to container 2. After the high humidity field is removed, the gripper returns to the original state and the foam is laid down. However, this kind of gripper can only grasp blocky objects, and it is difficult to grasp columnar objects. Therefore, a spiral gripper was designed inspired by vines. As shown in Figure 5 c, the gripper is similar to the programmable structures 1 and 2 in Figure 4, and can be twisted under the action of humidity. A complete grab process is shown in Video S2 and Figure 5 d, and a stick with a mass of 720 mg was grabbed by a gripper of 17 mg. First, the gripper is brought close to the stick and twisted under the action of high humidity to tightly wrap the stick. Then the stick is transferred from beaker 1 to beaker 2. Finally, the high humidity is removed, the gripper relaxed and the stick is put down. Besides, by mimicking the crawling process of an inchworm, a crawling robot was designed (Figure 5 e), which can move forward on a surface with unidirectional friction. The crawling process of the robot is shown in Video S3 and Figure 5 f. When the humidity increases, the curvature of the robot increases. Due to the directional friction characteristics of the contact surface, the front end of the robot does not move and the rear end moves forward. When the humidity is removed, the curvature of the robot decreases. At this time, the back end is anchored, and the front end moves forward. The above motions complete locomotion cycle. The crawling robot moves forward for 57 mm in 60 s, with an average speed of about 1 mm/s. With the increase of the driving frequency, the moving speed can be further increased. At present, the driving force of most humidity actuators is very small due to the small mass and low modulus, so the existing humidity-driven robots basically do not have good load-bearing capacity. Therefore, inspired by the weight-bearing behavior of the ants, we designed a bionic weight-bearing robot, whose structure is shown in Figure 5 g. With four deformable joints as the supporting legs and the middle part as the weight-bearing platform, a bearing robot of 180 mg can fully lift an object of 5 g under the action of humidity (Figure 5 h), or even barely lift objects of 10 g (Figure 5 i). The full process can be seen from Video S4. In the pre-deformed state (Figure 5 j), the robot can easily carry a load of 60 g. The structure does not tilt until the load reaches 80 g.

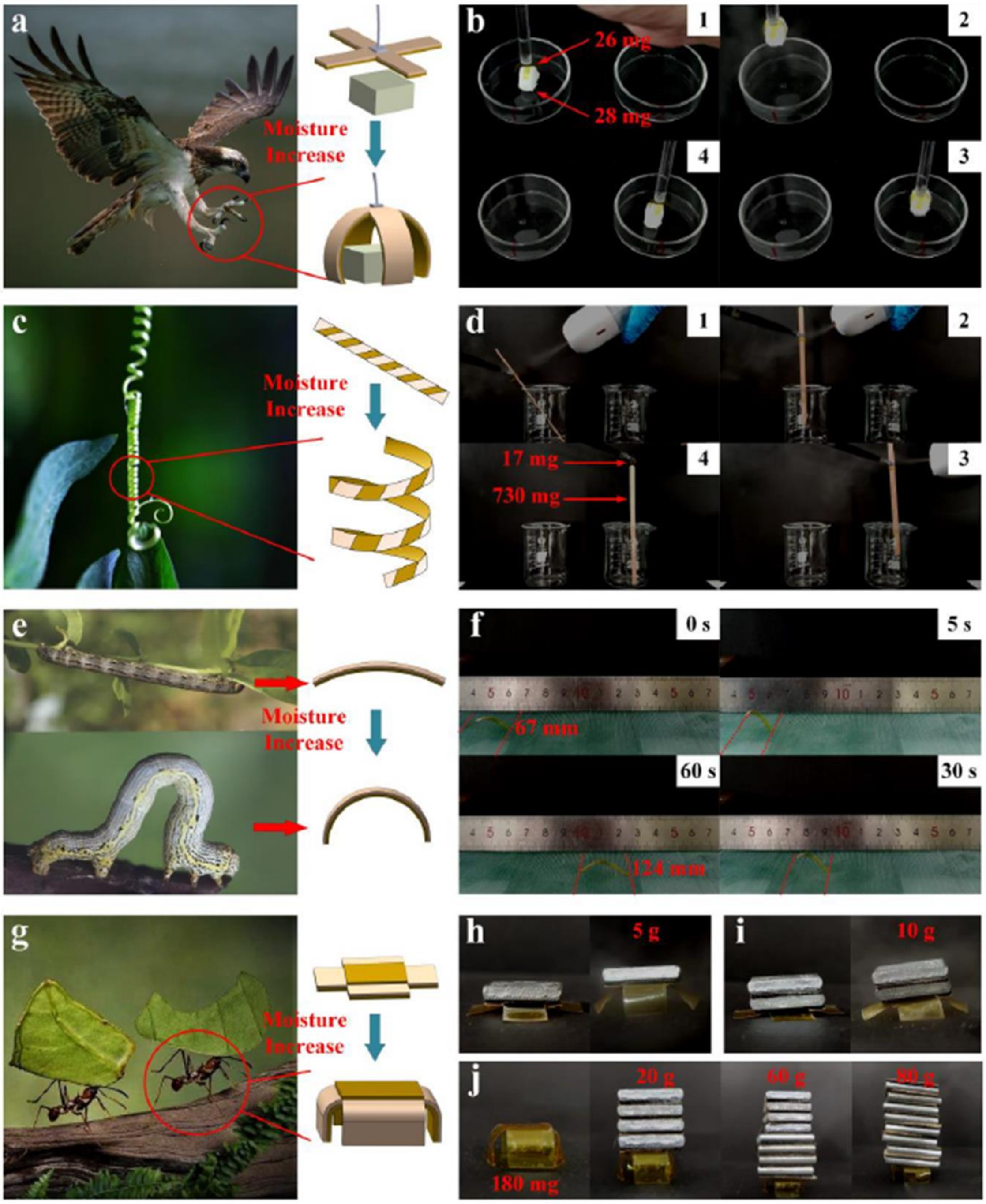

Figure 5. a. The cross-shaped flexible gripper inspired by the claws of birds. b. A typical grasping process using the cross-shaped gripper. c. The spiral gripper inspired by vines. d. A complete grab process using the

spiral gripper. e. The crawling robot inspired by inchworms. f. The crawling process of the robot. g. The bionic weight-bearing robot inspired by ants. h. The bearing robot lifts an object of 5 g. h. The bearing robot lifts an object of 10 g. j. The weight-bearing capacity of the robot in the pre-deformed state.

3.Conclusion

In this work, a facile and rapid method was proposed to prepare double-layer moisture-driven actuators with high performance, in which PI with good humidity insensitivity and thermal stability is selected as the binding layer, while Nafion is selected as the active layer for its high response speed and bending curvature compared with other humidity-sensitive materials. The prepared double-layer actuator has bidirectional deformation capability and good stability, the deformation range of which can be controlled by changing the thickness of the Nafion layer and the assembly humidity. Compared with other double-layer moisture-driven actuators, the prepared actuator not only has large bending curvature (from -0.98 cm^{-1} to 5.34 cm^{-1}) but also has high response speed (0.29 cm^{-1}/s). Through programmable structure design, a series of complex deformations were realized, including torsion, bending, "S" and "U" shaped deformation. By imitating organisms like birds, vines, inchworms and ants, a series of soft robots have been developed using the programmable structures to achieve behaviors including grasping, crawling, weight-bearing. The proposed preparing method with high speed and low cost and the realization of abundant functions for soft robots have shed light on the development of environmental-friendly functional microrobots obtaining energy supplement from ambient humidity.

4.Experiment Section

Materials: Nafion membranes with thickness of 0.051 (N-112), 0.89 (NE-1135) and 0.183 (N-117) mm were purchased from Dupont™ (USA). PI tape (RHL 12530#) was purchased from

RUNSEA Co., Ltd (Shenzhen, China). Auxiliary reagents including HCl, LiOH, NaOH, KOH, CH_3COOK, NaCl and allochroic silicagel were obtained from J&K Chemical Inc. (Beijing, China).

Preparation of single-layer actuator: The Nafion membrane was treated with ultrasonic cleaning (60°C, 60 min), acid boiling (HCl solution with a concentration of 2mol/L, 95°C, 30 min) and DI water boiling (95 °C, 30 min) in order. Then it was soaked in 0.2 mol/L alkali solution (LiOH, NaOH or KOH) for 2 h to conduct cations exchange.

Preparation of double-layer actuator: First, one side of the single-layer actuator was polished by sandblasting (sand-size of 220#, pressure of 0.4 MPa, time for 60s). Then the roughened surface was rinsed with deionized water to remove the sand. Finally, when the water on the roughened surface is volatilized, it was bonded with the PI tape.

Characterization and measurement: The cross section of the double-layer actuator was observed by ZEISS Sigma 500 field emission scanning electron microscope. The images and videos were captured by a digital camera (XIAOMI 10S). Changes in humidity are detected by a hygrometer (SMART SENSOR® AR837). The humidity during the bending curvature test of the double-layer actuators (the size of the cantilever structure is 5 mm × 25 mm) under a certain humidity is obtained by placing deionized water or allochroic silicagel in a closed box. Stable humidity of 22% RH and 74% RH in the stability test passed was obtained from the saturated salt solution of CH_3COOK and NaCl, respectively. The response speed of both single-layer actuators and double-layer actuators are tested by blowing air with saturated humidity through a tube with a diameter of 1 cm to the actuator of the cantilever (the size of the cantilever structure is 3 mm × 15 mm) using a wireless humidifier (HUMIDIFIER BP2). The crawling robot was driven by blowing air through a tube connected to the humidifier, and other robots were directly driven by the wireless humidifier.

Acknowledgements

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