Biodegradable Humidity Actuators for Sustainable Soft Robotics using Deliquescent Hydrogels

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***Abstract*— Intelligent materials offer new avenues when designing sustainable robotics as they allow for the creation of dynamic constructs which react autonomously to changes in the environment, such as humidity. Here we present a novel humidity actuator which exploits the unique property of deliquescent salts to allow for the spontaneous rehydration of hydrogels in ambient environments. By soaking a 2% w/v alginate, 3% w/v Agar composite in 1M calcium chloride, an intelligent humidity-driven actuator was developed. The hydrogel was able to gain 73.8±7.1% of its weight from a dehydrated state in just 6 hours through water absorption from ambient air. Using this novel formulation, linear and bilayer bending actuators were constructed. In addition to this, a biodegradable deliquescence-actuated artificial flower was demonstrated, highlighting this material's potential to act as an intelligent humidity actuator for the construction of environmentally-reactive biomimetic sustainable robotics.**

I. INTRODUCTION

The impact that non-disposable technologies have on the environment is becoming an increasingly detrimental and urgent problem [1]. This has driven scientists to seek green and sustainable solutions for soft robotic development. Intelligent materials [2] are materials imbued with unique chemistry which enable controlled deformation in response to a range of external stimuli such as light [3], temperature [4], pH [5], [6], magnetic fields [7] and electrical fields [8], [9]. By constructing robots from these smart materials, the ability to autonomously respond to the environment reduces the need not only for complex control electronics, but also internal energy sources. The development of sustainable smart materials which can be used to develop green soft robotics is therefore of great importance. One of the most promising materials which can be used to develop biodegradable soft robots are intelligent materials which respond to humidity.

Humidity is ubiquitous, dynamic and can act as a simple trigger to drive intelligent structures. A variety of different humidity-driven intelligent soft robots have been demonstrated such as grippers [10], [11], smart windows [12], sensors and biomimetic structures like seed drills [13], [14]. Furthermore, many of these unique robots have been designed to be biodegradable and cheap [10], [11] making them ideal for the design of sustainable robotics. However, humidity actuators typically require extremely thin dimensions [15], [16], [17] which although generating fast actuation responses, produce low forces. Furthermore, human intervention to initiate actuation, such as the use of humidity chambers for water supply is typically required [10], [11], [15]. For this reason, humidity triggered robotics have been limited to being paper thin and operating only in laboratory settings.

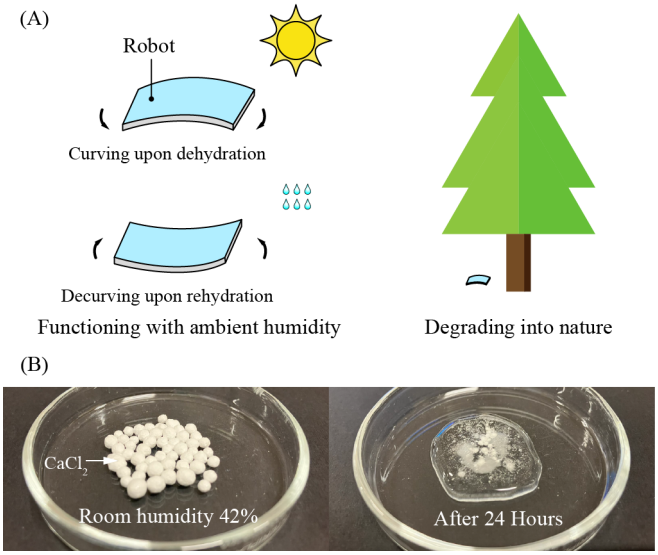

Fig. 1. (A) presents the concept of a sustainable humidity robot. (B) demonstrates the deliquescent effect of $CaCl_2$, where dehydrated $CaCl_2$ absorbed a large amount of water from ambient environment.

Calcium chloride is an abundant, sustainable salt used in a variety of applications from natural dehumidifiers [18], [19] to cooking [20], [21]. Moreover, calcium chloride displays a unique property known as deliquescence whereby as it is extremely hydroscopic, when left in an ambient environment it will absorb moisture from the atmosphere to such a degree that it dissolves in the absorbed water and forms a solution [22], Fig. 1 (B). We proposed that this property can be utilised to imbue edible, biodegradable hydrogels with significant humidity responsiveness.

Hydrogels are extremely hydrophilic polymers, able to absorb many times their own weight in water [23], [24], [25], [26]. These types of polymers are common in the natural environment and are used in a variety of fields

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such as tissue engineering[27], [28], contact lenses [29], wound dressing [30], 4D printing [4] and cooking [31], [32]. The incorporation of calcium chloride into a hydrogel biopolymer results in a synergistic combination, whereby the calcium chloride salt absorbs moisture from the environment inducing a rehydration of the hydrophilic polymer network. Correspondingly, this rehydration produces an expansion of the weight/volume of the polymer due to the influx of water. This influx of water will then produce a force which can be utilised to construct a variety of actuators. To evaluated the potential of this novel humidity-driven actuator, a composite of alginate/agar soaked in calcium chloride was constructed. The ability of this hydrogel to spontaneously absorb water from the ambient environment was evaluated, and its potential to construct sustainable biomimetic soft robots explored.

In this paper, we present the concept of a sustainable soft robot that can be powered by ambient humidity, as illustrated in Fig. 1 (A). The robot actuates (curls in this embodiment) and relaxes (decurls) in response to different moisture contents in the air. At the end of its lifecycle it degrades benignly in the natural environment. We developed a deliquescent hydrogel that can actively absorb water molecules from the environment without any human intervention, and demonstrated its application as bending and linear actuators.

II. MATERIALS, ACTUATORS, AND METHODS

A. Materials

Agar, alginic acid sodium salt from brown algae (alginate) and calcium chloride ($CaCl_2$) were all bought from Sigma-Aldrich. All experiments were conducted using deionized (DI) water. Biodegradable cellulose cloth was used as a backing layer to construct bending actuators. All materials were used as received, without further purification.

B. Preparation of Hydrogel Samples

Hydrogel samples were prepared with weight ratios according to table 1. Alginate was added to DI water and mixed at 500 rpm for 1 hour or until fully dissolved. Agar was then added and mixed at 90 °C for 1 hour at 500 rpm. For rehydration measurements hydrogel samples were cast into moulds of 2 mm high, 10 mm wide, and 50 mm long and left to cure for 20 minutes. Once cured, samples were soaked in solutions of either 0M, 1M, 3M or 5M $CaCl_2$ for 12 hours. Once samples had reached equilibrium with $CaCl_2$ they were dehydrated by placing them in an oven at 80 °C for 4 hours.

TABLE I
POLYMER WEIGHT RATIO OF DIFFERENT SAMPLES

Name	Agar (mg)	Alginate (mg)	DI water (mL)
0Alg3Agar	300	0	10
0.5Alg3Agar	300	50	10
1Alg3Agar	300	100	10
2Alg3Agar	300	200	10
1Alg1.5Agar	150	100	10

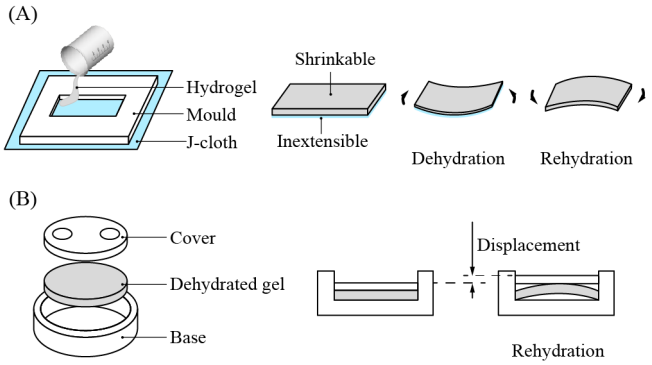

Fig. 2. Design and working principle of (A) the bending actuator and (B) the linear actuator.

C. Rehydration of Hydrogels

The rehydration of the samples were measured in the ambient environment of the climate monitored Bristol Robotics Laboratory at 23 °C and 40% relative humidity in average during the experiment. Rehydration was determined by measuring the percentage increase in weight of the samples after exposure to the ambient environment according to Eq. 1,

$$\frac{W_r - W_d}{W_d} \times 100 \tag{1}$$

where W_r is the rehydrated weight and W_d is the initial dehydrated weight. All experiments were preformed in triplicate.

D. Design of Actuators and Flower Demonstrator

The bending actuator was designed as a two-layer structure, as indicated in Fig. 2 (A). It was fabricated by casting the hydrogel into moulds of 3 mm high, 10 mm wide, 50 mm long with a biodegradable cloth (J-Cloth) and left to cure for 20 minutes. Because of the mismatch of the extensibility of the two layers, the actuator bends when the hydrogel layer shrinks upon dehydration, and uncurls when the hydrogel layer rehydrates and swells. A demonstration actuated flower was fabricated by installing three bending actuators, as petals, onto a 3D printed base object, acting as the peduncle.

As shown in Fig. 2 (B), the linear actuator consists of three parts, the base chamber, the dehydrated hydrogel and the cover. The hydrogel (15 mL) was first cast into a petri dish of diameter ϕ40 mm, then dehydrated as above, resulting in a dehydrated size of diameter ϕ35 mm and 2.5 mm thickness. It was then placed into the base chamber (ϕ35 mm) with the circular cover on top. When rehyhrated, the hydrogel swells and buckles upward due to the constraints of the chamber, inducing linear displacement of the cover.

E. Characterization of Bending, Linear Displacement and Flower Opening

The bending deformation of the bending actuators was characterized by using a high resolution camera placed at the side of the rehydrating actuators. The bending was captured periodically over 24 hours (once per hour during the first 12

hours and then at the end of the 24th hour). The percentage of curvature change was extracted by image processing techniques implemented in OpenCV in Python. The bending curvature was found by,

$$\kappa = \frac{1}{R} \tag{2}$$

where R is the radius of the characteristic circle, as illustrated in Fig. 6, and the percentage of curvature change was calculated by,

$$\frac{\kappa_r - \kappa_d}{\kappa_d} \times 100 \tag{3}$$

where κ_r is the rehydrated curvature and κ_d is the initial curvature after dehydration.

The linear displacement of the linear actuators was monitored by a laser distance sensor (Keyence LK-G152) for 24 hours. The data was recorded automatically in Matlab via a data acquisition device (NI DAQ USB-6211) at frequency of 10 Hz. The repeatability of the linear actuation was verified by repeating the dehydration, rehydration and distance measurement under same condition. All experiments were preformed in triplicate.

The opening process of the flower was monitored using two cameras (one side and the other overhead) for 5 hours.In the first 4 hours, it was under ambient humidity, and in the last hour, DI water was sprayed around the flow to simulate rainy weather in nature.

III. RESULTS AND DISCUSSION

Calcium chloride is an extremely hydrophilic salt that has the ability to absorb many times its own weight in water from the surrounding air [22]. This unique property has been exploited to demonstrate water harvesting polymers [33] but it's capacity as an actuator has yet to be evaluated, to the best of our knowledge. To determine the ability of this salt to imbue a hydrogel matrix with an intelligent response to humidity, alginate/agar composites containing $CaCl_2$ were fabricated and studied. The effects that the $CaCl_2$ concentration, polymer to water ratio, and polymer to polymer ratio had on the moisture absorption rate and capacity was evaluated.

A. Alginate/Agar Ratio Optimisation

Agar is a naturally occurring polysaccharide biopolymer derived from red algae. It is thermoplastic and was used to aid in the casting of the composites [34]. Alginate is also a naturally occurring biopolymer sourced from brown algae. It has a typical lifespan of 28 to 35 days [35] before it biodegrades and therefore it is estimated that the lifespan of our devices will be similar. Alginate is an anionic polysaccharide, crosslinked via the introduction of multivalent metal ions, such as calcium, which bond the carboxylic acid groups located on their guluronate residues [36]. It was thought that this interaction with the calcium ions would aid in the encapsulation of calcium chloride and the corresponding diffusion of water vapour into the polymer matrix. These two polymers were used to construct composites of different agar:alginate ratios which were then soaked in 5M calcium chloride. The agar concentration was fixed at 3% w/v Agar and the alginate concentrations ranged from 0%, 0.5%, 1% and 2% w/v, according to table 1. The effect that polymer ratios had on the rehydration of the polymer matrix was evaluated and the results shown in Fig. 3.

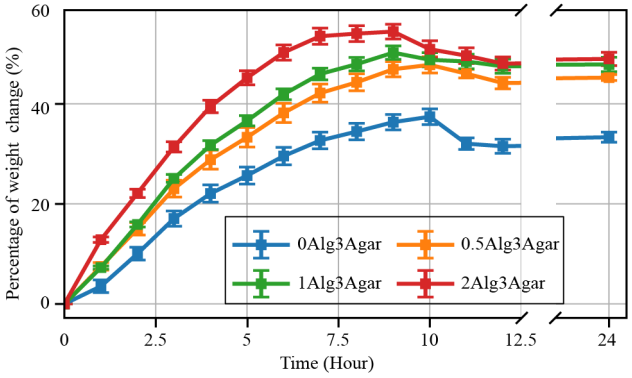

Fig. 3. The percentage recovery of weight during rehydration of samples after being soaked in 5M $CaCl_2$ for polymer to polymer ratio optimization.

It was found that although samples which did not contain alginate still display a hydroscopic weight gain from the contained calcium chloride, the inclusion of alginate significantly improved the absorbed water. This is illustrated in Fig. 3 with the increase in weight of the composite going from a 31.5±3.5% weight gain to a 48.1±2.6% weight gain after 12 hours exposure to the air for the 0Alg3Agar and 2Alg3Agar samples, respectively. In addition to this, not only did the total absorbed mass of water increase with an increase in alginate concentration, but the rate of water absorption also increased with larger quantities of alginate. This is illustrated by the 2Alg3Agar sample reaching its maximum weight after approximately 7 hours, while the 0Alg3Agar sample took 10 hours. This was proposed to be a result of the strong affinity of the alginate network for calcium ions. As alginate is known to form strong ionic bonds with calcium ions, it is thought that superior rehydration of the 2Alg3Agar samples is a result of the bound calcium ions in the polymer network leading to an increase in the density of calcium chloride within the composite. This correspondingly leads to an increased affinity for absorbed water vapour into the hydrogel matrix. Furthermore, from the superior performance of the 2Alg3Agar composite it can also be concluded that the optimal polymer ratio to be 2:3 alginate to agar.

As can be seen in Fig. 3, although the composites showed a clear increase in weight from the rehydration of the polymer matrix's from exposure to the ambient air, there was a clear decrease in the weight gain at approximately 10 hours. This was proposed to be a result of the humidity in the room not being stable and that the humidity decreased, resulting in the samples losing some gained water weight. Nevertheless, this demonstrates the dynamic response of these hydrogels to the surrounding environment, highlighting their novelty to act as intelligent humidity actuators for the construction of biomimetic sustainable robotics.

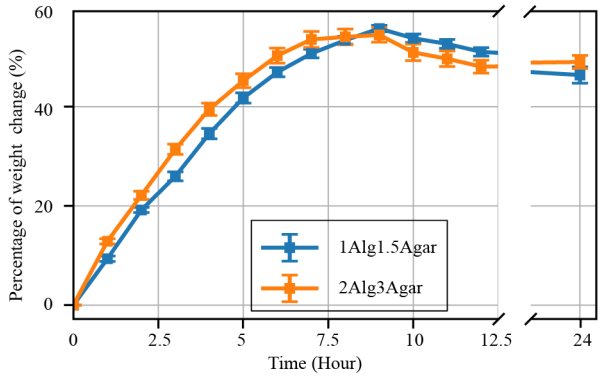

Fig. 4. The percentage recovery of weight during rehydration of samples after being soaked in 5M $CaCl_2$ for polymer to water ratio optimization.

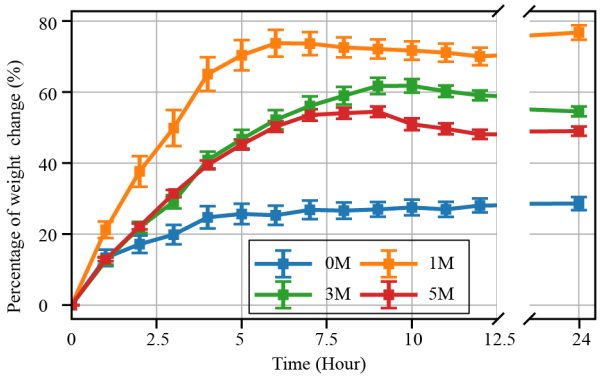

Fig. 5. The percentage recovery of weight during rehydration of 2Alg3Agar samples after being soaked in different $CaCl_2$ concentrations for polymer to salt ratio optimization.

B. Polymer/Water Ratio Optimisation

As discussed above, it was determined that the optimal polymer ratio was 2:3 alginate to agar. However, to assess the effect of polymer to water ratio on the capacity of the hydrogels to rehydrate, a study was conducted using a fixed polymer ratio of 2:3 alginate to agar with different polymer to water ratios. The polymer samples 2Alg3Agar and 1Alg1.5Agar were prepared according to table 1, soaked in 5M $CaCl_2$, dehydrated and left to rehydrate over the 24 hours. Samples with over 2% w/v alginate were not explored as the increased viscosity from the high concentration of alginate prevented accurate casting and processing. The results of this study are shown in Fig. 4.

From Fig. 4, it can be seen that the maximum rehydrated weight of the two samples were within the error of each other, implying that there was no real difference in the rehydration capacity of the two composites. These findings illustrate that the polymer to water ratio does not have a substantial effect on the total rehydration of the polymer network, compared to the effect that the polymer to polymer ratio has. However, it was found that the 2Alg3Agar sample had a faster rehydration rate when compared to the 1Alg1.5Agar sample as the 2Alg3Agar reached its maximum rehydration weight within 7 hours, whilst the 1Alg1.5Agar took 9 hours. Moreover, the 1Alg1.5Agar composite had a similar rehydration rate as the 1Alg3Agar composite, as can be seen in Fig. 3 and 4. These results indicate that although the polymer to polymer ratio has an effect on the maximum rehydration of the composites, the rate of rehydration is dependent on the concentration of alginate, whereby an increase in alginate concentration corresponds to an increased rehydration rate. From these results it can be seen that once again the 2Alg3Agar actuator is the optimal formula for the development of a intelligent biodegradable humidity actuator. Therefore, the 2Alg3Agar formulation was used to explore the effect of different calcium chloride concentrations on rehydration performance.

C. $CaCl_2$ Concentration Optimisation

Above, we optimised the polymer to polymer ratio and the polymer to water ratio to identify the optimal composite formula. We now consider the optimal concentration of $CaCl_2$. Using the 2Alg3Agar formula described in table 1, hydrogel samples were soaked in 4 different molar concentrations of $CaCl_2$; 0M, 1M, 3M and 5M. These samples were then dehydrated in an oven according to section II.B. and left to rehydrate over 24 hours. The results of the rehydration of the 2Alg3Agar gels soaked in different concentrations of $CaCl_2$ are shown in Fig. 5.

From the results in Fig. 5 it can clearly be seen that the samples infused with 1M $CaCl_2$ resulted in the best rehydration of the polymer, displaying a 73.8±7.1% weight gain in just 6 hours. Furthermore, the order of rehydration capacity for the different concentrations of $CaCl_2$ used was 1M>3M>5M>0M, highlighting the contribution of the $CaCl_2$ to the sustainable intelligent humidity actuator. It was first thought that a higher concentration of $CaCl_2$ would result in a larger rehydration, as more salt could absorb more water. However, the rehydration capacity was larger using lower concentrations of $CaCl_2$ (excluding 0M). This can be explained by considering the competing forces of polymer contraction due to the presence of the $CaCl_2$ and the swelling caused by the water absorbed through deliquescence of the $CaCl_2$. During hydration of the deliquescent $CaCl_2$, an expansive force from the absorbed water is produced within the hydrogel samples. Conversely, this expansive force simultaneously competes with the induced polymer collapse of the alginate network via the $CaCl_2$ promoting ionic crosslinking and functional group charge shielding. The samples with lower concentrations of $CaCl_2$ had a lower crosslinking density, and hence the polymers could expand to a greater volume and displayed superior rehydration. Furthermore, the samples soaked in 1M $CaCl_2$ also exhibited an improved rate of rehydration, reaching a maximum mass within just 6 hours. These findings demonstrate that the optimal formula to produce a sustainable intelligent humidity actuator is the 2Alg3Agar composite infused with 1M $CaCl_2$. This formula was used to construct the bilayer/linear actuators and artificial flower.

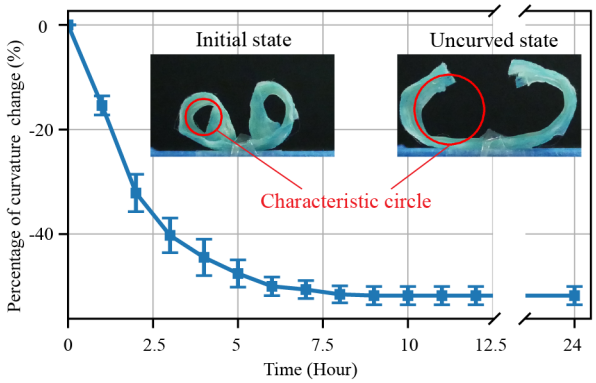

Fig. 6. The percentage curvature change of the bending actuator upon rehydration.

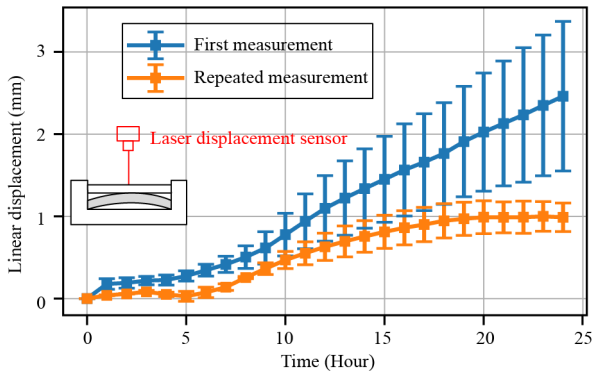

Fig. 7. Displacement of the linear actuator upon rehydration.

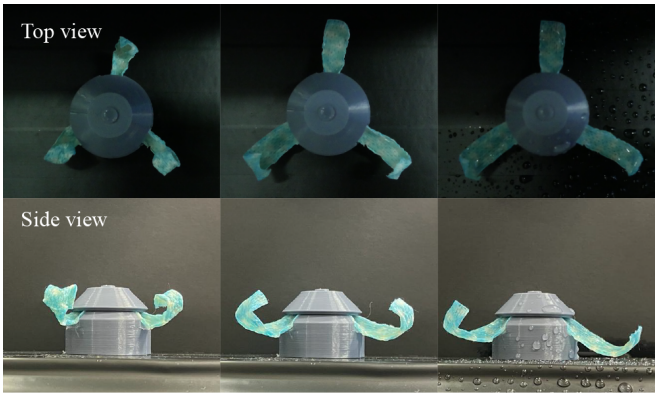

Fig. 8. Demonstration of an actuated flower constructed by three bending actuators.

D. Bending Actuator

A multi-layer bending actuator was designed to harness the asymmetric stretchability of materials upon stimulus. The actuator curled upon dehydration and uncurled once rehydrated. The insets of Fig. 6 show the initial state after dehydration and final state of rehydration. It is assumed that the hydrogel layer will bend homogeneously, so the actuator was observed to bent the most in the largest dimension while curling slightly in the other dimensions. The unbending capability due to the deliquescence effect was characterized by examining the bending curvature as detailed in Section II. The dehydrated actuator unbent at a fast speed during the initial two hours, then slowed gradually and stopped at the end of the 10th hour. As introduced earlier, the deliquescence behaviour of the hydrogel is a thermodynamic process that is affected by the ambient environment. The unbending behaviour of the actuator is affected by the deliquescence of the hydrogel and the mechanical property of the two constituent materials. This was likely the reason why the actuator did not flatten completely compared with the state before dehydration.

E. Linear Actuator

The linear actuator exploited the rehydration-induced swelling of a cylindrical hydrogel. Fig. 7 presents the linear displacement of the actuator over 24 hours for two cycles. The first cycle experienced an initially slow actuation followed by an increase in actuation rate, achieving a final upward displacement of approximately 2.4 mm (the thickness of the dehydrated hydrogel was approximately 2.5 mm). The second cycle presented a similar slow initial then faster movement, with an earlier plateau at around 20th hour. The inconsistency between the two cycles may be a result of the dehydration causing damage to the polymer network, preventing polymer rehydration.

F. Demonstration of an Actuated Flower

An actuated flower was demonstrated to show a potential application of the deliquescent hydrogel towards sustainable soft robots. As shown in Fig. 8, the initially curled flower gradually opened when stimulated by ambient humidity during the first 4 hours, following the deformation trend of the bending actuators in Fig. 6. At the end of the 4th hour, natural rainy weather was simulated by spraying DI water around the flower. The flower was observed to further open at a faster speed. This also illustrated that the system does not degrade in rain.

IV. CONCLUSIONS

Smart materials offer researchers new avenues to design sustainable robotics as they allow for the creation of dynamic constructs which respond autonomously to the environment. This ability of environmental reactivity removes the need for complex electronic control components and integrated power sources. To further the state-of-the-art of this field, a smart humidity actuator was developed which exploited the deliquescent properties of calcium chloride to imbue a hydrogel composite of agar/alginate with the ability to rehydrate in an ambient environment. Using a 2% w/v alginate and 3% w/v alginate composite soaked in 1M calcium chloride, an autonomous humidity actuator was developed which was able to gain 73.8±7.1% of its weight from a dehydrated state in just 6 hours from the ambient environment. Using this novel smart humidity actuator, linear and bending actuators were developed. Lastly, a biodegradable artificial flower was

constructed using this novel formulation which was able to spontaneously bloom, illustrating the potential for the development of biomimetic soft robotics. In this work we have presented a new form of smart humidity actuator which is not constricted to paper thin dimensions and does not require human intervention to display an actuation response, highlighting the novelty of this formulation to further the development of environmentally reactive and sustainable robotics.

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